A femtosecond journey from dream to nightmare:

Time evolution of reduced density matrices using BBGKY hierarchy

Aalto University

M.J. Hashemi [1,2], A. Akbari [3], A. Rubio [3], R. Nieminen [1] and R. van Leeuwen [4]

[1] COMP, Department of Applied Physics, Aalto University, Finland.

[2] Department of Physics, University of Helsinki, Finland.

[3] Departamento de Física de Materiales, Universidad del País Vasco, San Sebastián, Spain.

[4] Department of Physics, Nanoscience Center, University of Jyväskylä, Finland.

Introduction

- The equation of motion for the full density matrix of a system can be rewritten as a hierarchy of equations that describes the time evolution of each reduced density matrix (RDM). This is called ***BBGKY*** hierarchy.

Compatibility

$$
\begin{aligned}
i\partial_t \gamma &= A_1 \gamma + S_1(\Gamma^{(2)}) \\
i\partial_t \Gamma^{(2)} &= A_2 \Gamma^{(2)} + S_2(\Gamma^{(3)}) \\
i\partial_t \Gamma^{(3)} &= A_3 \Gamma^{(3)} + S_3(\Gamma^{(4)}) \\
i\partial_t \Gamma^{(4)} &= A_4 \Gamma^{(4)} + S_4(\Gamma^{(5)}) \\
&\mathit{etc.}
\end{aligned}
$$

- If we truncate the hierarchy at the first level we get the time-dependent reduced density matrix functional theory (TDRDMFT).

- But here, we truncate the hierarchy at the second level to obtain more accurate results.

- Thus, we need to approximate $\mathbf{\Gamma^{(3)}}$ as a functional of $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$.

- The easiest approximation is to put, $\mathbf{\Gamma^{(3)} = 0}$. We call that 3-body collision integral free *(3b-CIF)* approximation.

- We can also construct $\mathbf{\Gamma^{(3)}}$ as a slater determinant of $\boldsymbol{\gamma}$. That is called 3-body non-interacting approximation *(3b-NIA)* and its mathematical form is:

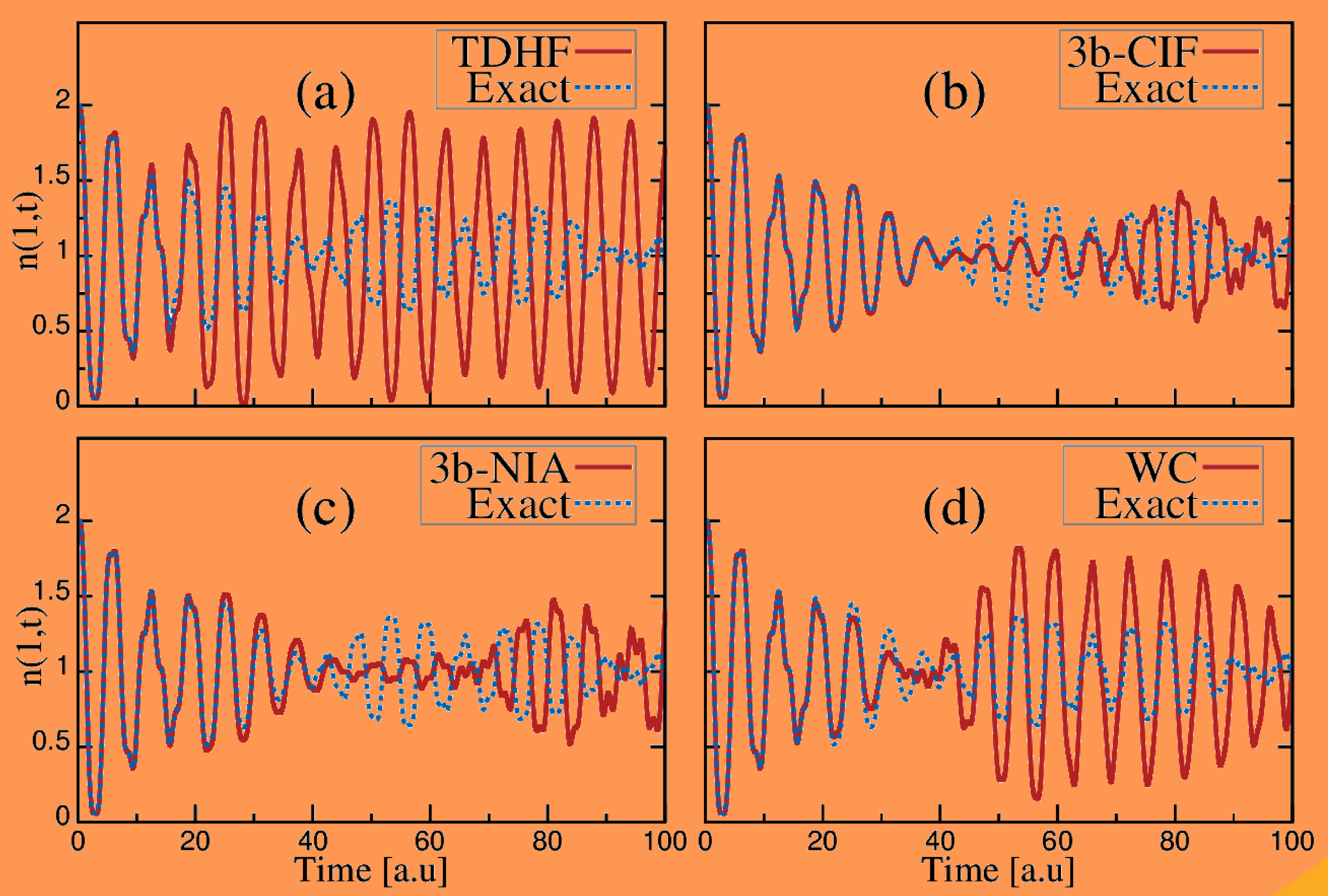
$$
\Gamma^{(3)}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3;\mathbf{x}'_1\mathbf{x}'_2\mathbf{x}'_3,t) = \begin{vmatrix} \gamma(\mathbf{x}_1\mathbf{x}'_1,t) & \gamma(\mathbf{x}_1\mathbf{x}'_2,t) & \gamma(\mathbf{x}_1\mathbf{x}'_3,t) \\ \gamma(\mathbf{x}_2\mathbf{x}'_1,t) & \gamma(\mathbf{x}_2\mathbf{x}'_2,t) & \gamma(\mathbf{x}_2\mathbf{x}'_3,t) \\ \gamma(\mathbf{x}_3\mathbf{x}'_1,t) & \gamma(\mathbf{x}_3\mathbf{x}'_2,t) & \gamma(\mathbf{x}_3\mathbf{x}'_3,t) \end{vmatrix}
$$

- A more advanced approximation consists of an antisymmetrized product of $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$, as well. We call that *WC* approximation.

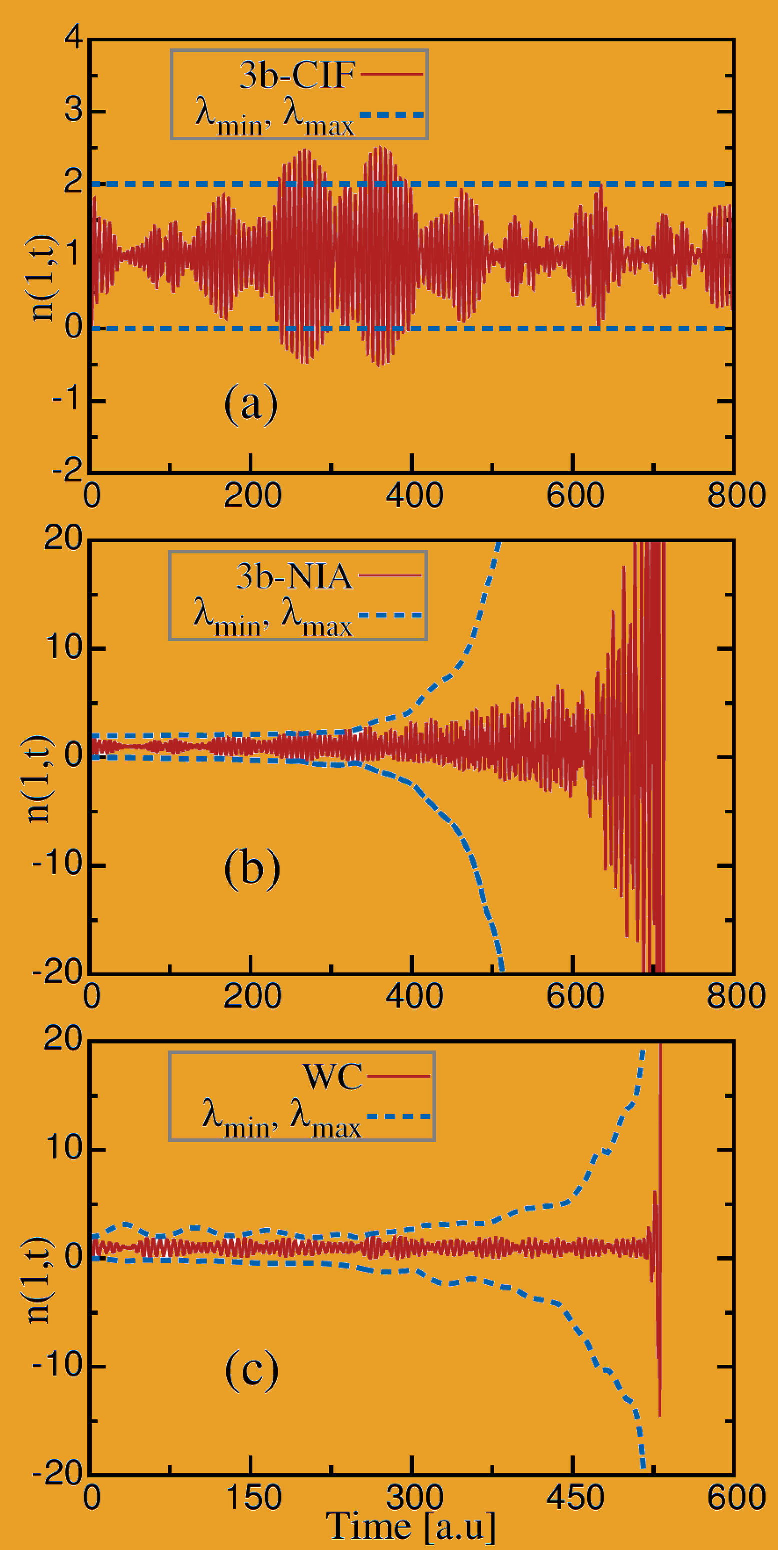
$$
\Gamma^{(3)}_{\mathrm{WC}} = -12\,\gamma\wedge\gamma\wedge\gamma + 9\,\gamma\wedge\Gamma
$$

Results

- Our test system is a four-site Hubbard chain with four electrons. The electrons initially filled the two left-most sites. Hopping parameter is set to unity and the on-site potential is *0.1*. Here, *n(1,t)* is the electronic density in site *1*.



- But if we propagate it for longer time, the electronic density will either violate the frmionic inequality or even diverges.



Where is the problem?

- When we approximate $\mathbf{\Gamma^{(3)}}$, the link between the first and the second equation (compatibility) does not hold any longer.

- On the other hand, reduced density matrices need to be positive-semidefinite at all times and some of the approximations are not built well enough to keep this property.

- If an approximation retains both of these properties, it will solve the problem of divergence and acquiring negative densities.

$\Gamma^{(3)}$ approximations	Compatibility of Equations	Positive-Semidefiniteness of Approximations	Violating ☹ Diverging ☠
$3b-CIF$	×	×	☹
$3b-NIA$	×	×	☠
WC	×	×	☠
Compatible (1)	✓	✓	☹
Compatible (2)	✓	×	☠
$\left(\frac{N-2}{N}\right)\gamma\,\Gamma$	×	×	☹
$3b-NIA$, WC only second equation	Does not Matter	×	☠
$\left(\frac{N-2}{N}\right)\gamma\,\Gamma$ only second equation	Does not Matter	✓	☹

More?: *PHYSICAL REVIEW B 85, 235121 (2012)*